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ABSTRACT

Understanding Interfacial Reactions at the Electrode/Electrolyte Interface using Neural Network Potential Molecular Dynamics

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First-principles calculations based on quantum mechanics can accurately reproduce the crystal and electronic structures of inorganic solids. However, their high computational cost limits the number of atoms that can be handled, making simulations time-consuming. Recently, Neural Network Potentials (NNP), trained on extensive datasets of first-principles calculations, have enabled efficient and accurate energy and force predictions for a wide range of materials. In this study, we investigate the application of NNP to lithium-ion battery interfacial reactions, which significantly impact battery life and charge-discharge performance but remain difficult to analyze. Molecular dynamics simulations reveal charge separation at the interface, where Li+ ions accumulate in the solid electrolyte while electrons concentrate in the metal. Analysis of Li defect formation energy suggests deviations near the interface, indicating changes in lithium chemical potential. Furthermore, Density Functional Theory (DFT) calculations performed on NNP-MD derived structures reveal that the solid electrolyte exhibits insulating properties with a band gap of approximately 4.5 eV. Band bending at the interface leads to the formation of a space-charge layer due to excess Li ions, which suppresses electron exchange between the electrolyte and metal. This phenomenon highlights the importance of controlling band bending for optimizing the performance of all-solid-state lithium batteries, emphasizing the role of interface engineering in next-generation battery development.

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